Ferromagnetic Resonance Studies in Some Polycrystalline Magnesium Nickel Ferrites

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Ferromagnetic resonance measurements were carried out on various compositions of magnesium nickel ferrites prepared by various heat treatments. Corrections were applied for polycrystalline and size effects for calculating the g factor and linewidth. The variation of these parameters has been explained on the basis of the cation distribution in these compounds as obtained from magnetization data assuming a collinear model of spin distribution.

Introduction

A clear understanding of the phenomenon of ferromagnetic resonance in ferrites has come about after the concept of two interacting sublattices of different magnetizations and g factors was extended to resonance by Kittel (1) and Wangsness (2). Though a lot of published data are available on the ferromagnetic resonance studies on ferrites, some magnesium-containing ferrites have not been systematically investigated because the cation distribution in such systems is very sensitive to preparative conditions. Magnesium nickel ferrites are interesting because the magnetic properties of the various compositions subjected to different heat treatments have to be related to the redistribution of Mg^{2+} and Fe^{3+} ion over the A and B sites, since Ni^{2+} occupies only the B sites. The aim of the present study is to understand the effect of cation distributions on the FMR parameters g_{eff} and linewidth (ΔH) in this system. Appropriate corrections for polycrystalline and size effects were made to obtain reliable values of the above parameters.

Experimental

Monophasic compositions of the ferrites $Ni_xMg_{1-x}Fe_2O_4$, where $0 \leq x \leq 1$, were prepared by standard ceramic techniques using appropriate amounts of NiO, MgO, and α-Fe_2O_3 of high purity and heating at 1575 K for 3 hr. One set of compounds was quenched from 1575 K to room temperature and another was furnace-cooled at the rate of about 60 K per hour. The packing densities of the sintered compacts were above 93% of their X-ray densities. The saturation magnetization ($4\pi M_s$) has been measured at various temperatures and extrapolated to 0 K to determine the cation distributions (3).

For FMR measurements spherical samples were made by an adaptation of Bond's technique (4) and placed on the base of a TE_{116}-type cavity (5) at a frequency of 9.94 GHz (superinvar cavity-stabilized Gunn di-

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TABLE I

STRUCTURAL AND MAGNETIC PROPERTIES OF THE SYSTEM $Ni_xMg_{1-x}Fe_2O_4$

	Furnace-cooled samples				Quenched samples			
		$4\pi M_s$ (Gauss)		$Mg^{2+}_{A\,site}$		$4\pi M_s$ (Gauss)		$Mg^{2+}_{A\,site}$
Composition (x)	a (Å)	300 K	0 K[a]	$\alpha(\pm 0.002)$	a (Å)	300 K	0 K[a]	$\alpha(\pm 0.002)$
0.0	8.372	1335	1755	0.120	8.400	3005	3807	0.225
0.2	8.350	1480	1700	0.072	8.368	2916	3468	0.190
0.4	8.362	1700	1952	0.047	8.354	2814	3418	0.159
0.5	8.341	1890	2091	0.034	8.351	2796	3066	0.114
0.6	8.355	1945	2288	0.035	8.350	2815	3116	0.100
0.8	8.333	2350	2886	0.035	8.332	2994	3264	0.060
1.0	8.320	3050	3378	—	8.320	3111	3382	—

[a] Obtained by extrapolating the $4\pi M_s$ vs T curve.

ode) at room temperature. The diameter dependence of the resonance field was studied in order to obtain g_{eff} values within 1% accuracy. The factors g_{eff} and ΔH were measured for both quenched and furnace-cooled samples.

Results and Discussion

The structural and magnetic properties of the ferrites are summarized in Table I. These properties have been discussed elsewhere (3). The cation distributions in these compounds can, in general, be written as

$$Mg^{2+}_\alpha Fe^{3+}_{1-\alpha}[Ni^{2+}_{1-(\alpha+\beta)}Mg^{2+}_\beta Fe^{3+}_{1+\alpha}]O^{2-}_4, \quad (1)$$
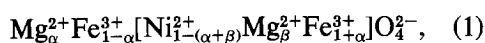
where $\alpha + \beta = 1 - x$. The value of α in the above expression can be written as

$$\alpha = \frac{n_B - x\mu_{Ni^{2+}}}{2\mu_{Fe^{3+}}(1-x)}, \quad (2)$$

where n_B is the saturation magnetic moment in Bohr magnetons at 0 K and $\mu_{Ni^{2+}}$ and $\mu_{Fe^{3+}}$ are 2.3 and 5.0 μ_B, respectively (5). From the value of α (Table I), and using Eq. (1), the cation distributions for all compositions could be obtained. The effective g factor can be written as (6)

$$g_{eff} = \sum_K M_K \bigg/ \sum_K \left(M_K \bigg/ g_K\right), \quad (3)$$

where M_K are the sublattice magnetizations and g_K the corresponding g factors. Substituting for M_K from the cation distributions and taking g_K as 2.3 and 5 for Ni^{2+} and Fe^{3+}, respectively,

$$g_{eff} = 2\left[\frac{2.3(1-(\alpha+\beta)) + 10\alpha}{2(1-(\alpha+\beta)) + 10\alpha}\right]. \quad (4)$$

This assumes that Ni^{2+} occupies the B site alone due to its strong octahedral site preference (7) and that the spins are aligned collinearly, i.e., there is no canting. This assumption arises from the fact that Mössbauer spectral studies (8) and neutron diffraction studies (9) on pure magnesium ferrite and nickel ferrite have shown them to be collinear systems. There is no mention in the literature of studies on nickel–magnesium ferrites that suggest a noncollinearity of spins. In the course of the present investigation, the validity of the assumption that the spin arrangement is collinear is tested. The g_{eff} values thus calculated are given in Tables II and III.

The ferromagnetic resonance curves for two typical samples are given in Fig. 1. For calculating g_{eff} and ΔH of these compounds, however, polycrystalline and size effects have to be considered. In the case of poly-

<div align="center">TABLE II</div>

<div align="center">FERROMAGNETIC RESONANCE DATA FOR THE SYSTEM $Ni_xMg_{1-x}Fe_2O_4$ (FURNACE COOLED)</div>

Composition (x)	Porosity (p)	$\Delta H \pm 10$ Oe (for sphere diameter of 0.01 cm)	$H_0 \pm 20$ Oe	$K_1/2M$ (Oe)	$p4\pi M_s/6$ (Oe)	g_{eff} (± 0.02)	g_{eff} (from cation distribution)
0.0	0.076	530	3230	94	17	2.02	2.00
0.2	0.070	645	3060	110	18	2.12	2.11
0.4	0.062	680	2950	119	18	2.19	2.19
0.5	0.080	705	2880	117	25	2.23	2.22
0.6	0.097	700	2870	124	32	2.24	2.23
0.8	0.061	595	2840	119	24	2.26	2.25
1.0	0.070	450	2790	114	36	2.30	2.30

crystalline magnetic insulators there are various factors which contribute to the resonance field. If a spherical sample is placed at the base of a cavity, the resonance frequency (ω_r) is given by (10)

$$\omega_r = \gamma(H_{ext} + H_a + H_p + H_e + H_{id}), \quad (5)$$

where the subscripts stand for: ext, externally applied; a, anisotropy; p, porosity; e, eddy currents within the cavity wall; and id, inhomogeneous demagnetizations. The corrections for these follow.

Schlomann (11) calculated the effect of the random orientation of crystallites in a polycrystalline sample based on an idealized model and found that the dipole sees an additional field given by

$$H_a = \left| \frac{K_1}{2M_s} \right|. \quad (6)$$

The values of K_1 for this system have been obtained from the literature (12, 13). Secondly, the demagnetizing effect of the pores at the grain boundaries also shifts the resonance field. The expression for this, assuming pores of ellipsoidal shape, is given by (14)

$$H_p = (1/6)p4\pi M_s. \quad (7)$$

Finally, the size effects, i.e., (a) the induction of eddy current on the walls of the cavity due to the precession of dipoles in the samples when placed in the cavity and (b) the inhomogeneous demagnetizing field

<div align="center">TABLE III</div>

<div align="center">FERROMAGNETIC RESONANCE DATA FOR THE SYSTEM $Ni_xMg_{1-x}Fe_2O_4$ (QUENCHED)</div>

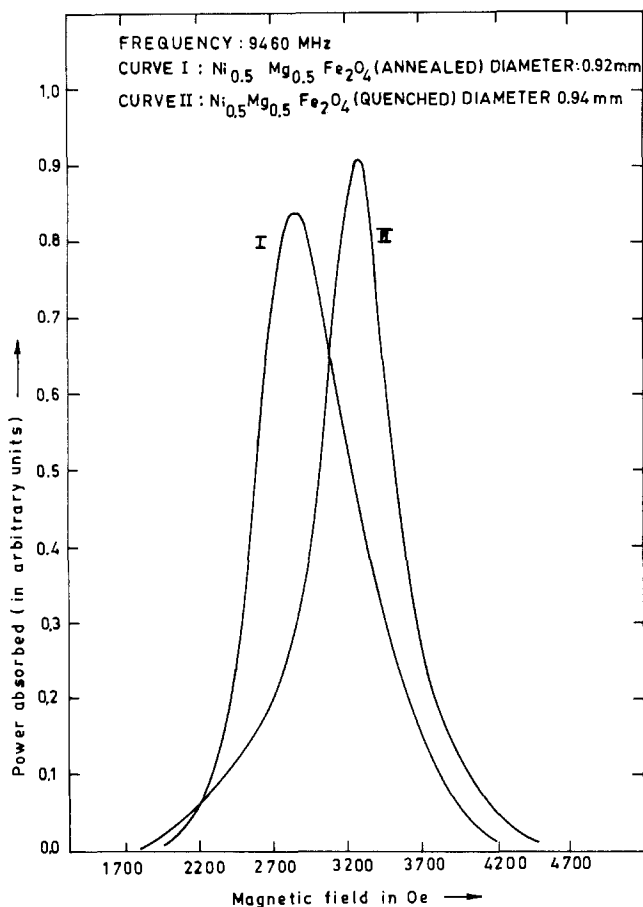| Composition (x)	Porosity (p)	$\Delta H \pm 10$ Oe (for sphere diameter of 0.01 cm)	$H_0 \pm 20$ Oe	$K_1/2M$ (Oe)	$p4\pi M_s/6$ (Oe)	g_{eff} (± 0.02)	g_{eff} (from cation distribution)
0.0	0.047	586	3250	84	24	2.01	2.00
0.2	0.063	527	3180	91	30	2.05	2.05
0.4	0.053	512	3080	99	26	2.11	2.10
0.5	0.054	527	3040	102	25	2.13	2.14
0.6	0.056	552	2980	104	22	2.17	2.16
0.8	0.057	577	2910	102	24	2.22	2.22
1.0	0.075	570	2800	103	39	2.30	2.30

FIG. 1. Ferromagnetic resonance curves for two representative samples.

over the sample, lead to additional shifts in the resonance field (15). Since H_e and H_{id} are dependent on the diameter of the sample, the resonance field was, therefore, measured as a function of sphere diameter and extrapolated (15) to give a diameter-independent resonance field (H_0) automatically corrected for H_e and H_{id}. The values of H_a, H_p, and the g_{eff} thus calculated are given in Tables II and III for the furnace-cooled and quenched samples, respectively.

There is excellent agreement between the g_{eff} values obtained directly from FMR measurements and those calculated from the cation distributions obtained from mag-netization studies (Fig. 2). This excellent agreement of g_{eff} data justifies the assumption about the collinear arrangement of spins.

In the case of magnesium ferrite the effective g factor is the same for furnace-cooled as well as quenched samples even though there is a large change in the cation distribution. This is because the only magnetic ion present, Fe^{3+}, though rearranged, shows the same g factor at A and B sites due to its spherical ground state. As reported earlier (3), the heat treatment does not change the cation distribution in nickel ferrite and hence it shows the same value of g_{eff} for furnace-cooled and quenched sam-
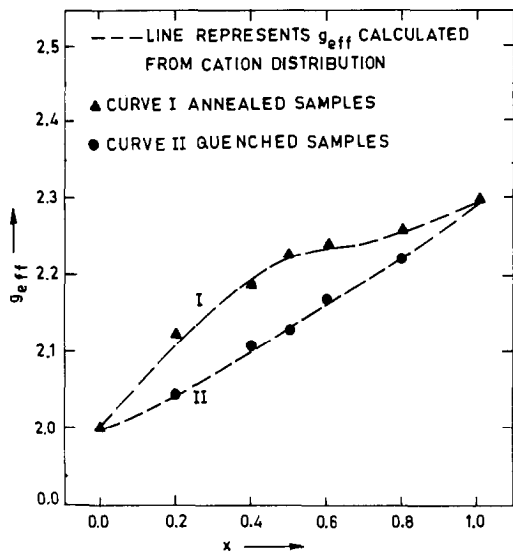
FIG. 2. Variation of g_{eff} with composition for the system $Ni_xMg_{1-x}Fe_2O_4$.

ples. In the intermediate compositions, however, the difference in the distribution of Mg^{2+} and Fe^{3+} over the A and B sites leads to difference in the g_{eff} values observed for furnace-cooled and quenched compositions.

Figure 3 represents the variation of ΔH with composition for spheres of diameter
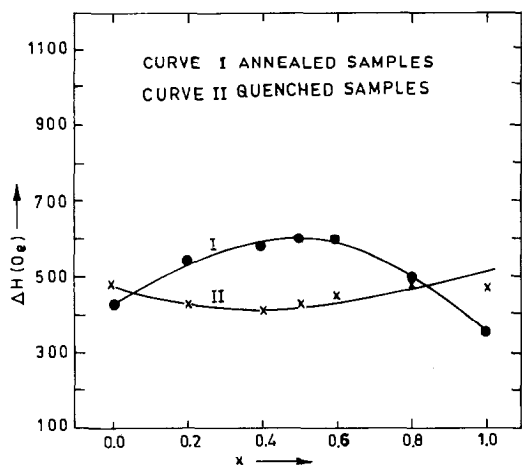
~ 1 mm. In the case of end compositions ($x = 0, 1.0$) the furnace-cooled samples have lower ΔH values, probably because they represent a situation where the ions have been allowed to come to an equilibrium and there is more regularity in arrangement. The maximum at $x = 0.5$ shown by furnace-cooled samples is difficult to explain from the present set of studies. The behavior of ΔH remained the same even after applying corrections for polycrystalline effects and a proper understanding would require the study of ΔH as a function of temperature.

Conclusions

The following conclusions may be drawn from the above studies:

(a) The calculation of FMR g_{eff} factors requires a proper understanding of internal fields, and both polycrystalline and size effects must be properly accounted for to obtain the true g_{eff} values.

(b) The g_{eff} behavior can be accounted for by considering a two-sublattice model and by employing Eq. (3).

(c) The variation of g_{eff} with composition is in very good agreement with the cation distributions as deduced from magnetization measurements, assuming a collinear model of spin arrangement.

(d) It is difficult to explain the variation of linewidth with composition from room-temperature values alone.

FIG. 3. Variation of ΔH with composition for the system $Ni_xMg_{1-x}Fe_2O_4$.

References

1. C. KITTEL, Phys. Rev. **82**, 565 (1951).
2. R. K. WANGSNESS, Phys. Rev. **93**, 68 (1954).
3. K. SESHAN, A. L. SHASHIMOHAN, D. K. CHAKRABARTY, AND A. B. BISWAS, Phys. Status Solidi A **68**, 97 (1981).
4. W. L. BOND, Rev. Sci. Instrum. **25**, 401 (1954).
5. M. J. PATNI, Ph.D. thesis, Indian Institute of Technology, Bombay (1972).

6. G. T. RADO AND H. SUHL, "Magnetism I," Academic Press, New York (1963).

7. A. MILLER, *J. Appl. Phys.* **30**(4), 245 (1959).

8. G. A. SAWATZKY, F. VAN DER WOUDE, AND A. H. MORRISH, *Phys. Rev.* **187**(2), 747 (1969).

9. E. WEISER, V. A. PROVITSKII, E. F. MAKAROV, AND K. KLEINSTUCK, *Phys. Status Solidi A* **25**, 607 (1968).

10. C. M. SRIVASTAVA AND M. J. PATNI, "Ferrites" (Y. Hoshino, S. Iida, and M. Sujimoto, Eds.), Univ. of Tokyo Press, Tokyo (1973).

11. E. SCHLOMANN, "Conference on Magnetism and Magnetic Materials," *AIE Spec. Publ.* **91**, 600 (1956).

12. C. J. KREISSMAN AND S. E. HARRISON, *Phys. Rev.* **103**, 857 (1958).

13. W. H. VON AULOCK, "Handbook of Microwave Ferrite Materials," Academic Press, New York (1965).

14. C. M. SRIVASTAVA, M. J. PATNI, AND N. G. NANADIKAR, *J. Phys.* **38**(4), 267 (1977).

15. M. J. PATNI AND C. M. SRIVASTAVA, "Ferrites" (Y. Hoshino, S. Iida, and M. Sujimoto, Eds.), Univ. of Tokyo Press, Tokyo (1971).